



# DECLARATION

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This is March 22, 2006.

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JAPAN PATENT OFFICE

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[Name of the Invention] VISCOSITY INDEX IMPROVER AND LUBE OIL  
COMPOSITION

[Claims]

5    [Claim 1]

        A viscosity index improver, which comprises a copolymer  
(A) comprising, as essential constituent monomers, a monomer  
(a) represented by the following general formula (1) and a  
monomer (h) which contains a hydroxyl group:

10    General formula

[Chemical formula 1]



15    wherein  $\text{R}^1$  is a hydrogen atom or a methyl group, X is an  
alkylene group containing 2-4 carbon atoms, n is 0 or an  
integer of 1-20, and  $\text{R}^2$  is a branched alkyl group containing  
20-36 carbon atoms.

[Claim 2]

20          The viscosity index improver according to claim 1,  
wherein the copolymer (A) further comprises, as an essential  
constituent monomer, at least one monomer selected from the  
group consisting of an alkyl (meth)acrylate (b) in which an  
alkyl group contains 1-4 carbon atoms, and an alkyl  
25    (meth)acrylates (c) in which an alkyl group contains 8-18  
carbon atoms or a straight-chain alkyl group contains 20-24

carbon atoms.

[Claim 3]

The viscosity index improver according to claim 2, wherein the copolymer (A) comprises 5-90 wt% of the monomer (a), 5-50 wt% of the monomer (h), and 5-90 wt% of at least one monomer selected from the group consisting of the monomer (b) and the monomer (c), relative to the weight of the copolymer (A).

[Claim 4]

The viscosity index improver according to any one of claims 1-3, wherein the monomer (a) constituting the copolymer (A) is a monomer represented by the general formula (1) in which n is 0.

[Claim 5]

The viscosity index improver according to claim 4, wherein the monomer (a) constituting the copolymer (A) is 2-decyltetradecyl (meth)acrylate.

[Claim 6]

The viscosity index improver according to any one of claims 1-5, wherein the copolymer (A) has a weight-average molecular weight of 5,000-35,000.

[Claim 7]

An additive composition for lube oil, which comprises the viscosity index improver according to any one of claims 1-6, and at least one selected from the group consisting of

a diluent and other additives.

[Claim 8]

The additive composition for lube oil according to claim 8, wherein the amount of the diluent is 5-900 wt%,  
5 relative to the weight of the viscosity index improver.

[Claim 9]

The additive composition for lube oil according to claim 7 or 8, which further comprises, as other additive, 0.001-10 wt% of a (meth)acrylate copolymer (B) other than the  
10 copolymer (A), relative to the weight of the copolymer (A).

[Claim 10]

A lube oil composition, which comprises the viscosity index improver according to any one of claims 1-6, and base oil.

15 [Claim 11]

A lube oil composition, which comprises the additive composition for lube oil according to any one of claims 7-10, and base oil.

[Claim 12]

20 The lube oil composition according to claim 10 or 11, wherein the base oil has a kinematic viscosity of 1-18 mm<sup>2</sup>/s at 100°C and a viscosity index of at least 60.

[Detailed Description of the Invention]

[0001]

25 [Field of the Invention]

This invention relates to a viscosity index improver, an additive composition for lube oil, and a lube oil composition containing them. More particularly, this invention relates to the viscosity index improver, the additive composition for lube oil, which are excellent in viscosity-index-improvement ability, and the lube oil composition containing them.

[0002]

[Description of the Prior Art]

10 With the recent tendency of global environmental protection, there is being an increased demand much more improvement in fuel-efficiency of automobiles. As one means of improving such fuel-efficiency, reduction of friction loss by making viscosity of lube oil lower is mentioned. However, 15 if only the viscosity of lube oil is made lower, the problems of spill of lube oil and seizure arise. In order to solve these problems, it is generally needed to raise the viscosity index of lube oil and viscosity index improvers comprising various kinds of polymethacrylate copolymers have been 20 proposed from the former (for example, JP7-48421A, JP7-62372A).

[0003]

[Problem(s) to be solved by the Invention]

25 However, the conventional polymethacrylate viscosity index improvers have problems that they are inadequate not

only in viscosity-index-improvement effectiveness but also in shear stability.

[0004]

[Means for Solving the Problem]

5           We, the present inventors have assiduously studied and have found out a polymer comprising specific monomers excellent in viscosity-index-improvement ability and shear stability. On the basis this finding, we have completed this invention.

10           The present invention relates to a viscosity index improver, which comprises a copolymer (A) comprising, as essential constituent monomers, a monomer (a) represented by the following general formula (1) and a monomer (h) which contains a hydroxyl group; an additive composition for lube  
15 oil, which comprises the above-described viscosity index improver, and at least one selected from the group consisting of a diluent and other additives; a lube oil composition, which comprises the above-described viscosity index improver and base oil; and a lube oil composition, which comprises the  
20 above-described additive composition for lube oil and base oil:

General formula

[0005]



[Chemical formula 2]



5 [0006]

wherein  $\text{R}^1$  is a hydrogen atom or a methyl group, X is an alkylene group containing 2-4 carbon atoms, n is 0 or an integer of 1-20, and  $\text{R}^2$  is a branched alkyl group containing 20-36 carbon atoms.

10 [0007]

[Embodiment of the Invention]

In the monomer (a) represented by the general formula (1) of this invention,  $\text{R}^2$  is a branched alkyl group containing 20-36 carbon atoms, and is concretely classified by the position of the branch, as described below.

(1) 1-alkyl groups (alkyl groups containing a branch at the first place): 1-ethyloctadecyl group, 1-hexyltetradecyl group, 1-octyldodecyl group, 1-butyleicocyl group, 1-hexyloctadecyl group, 1-octylhexadecyl group, 1-decyltetradecyl group, 1-undesyltridecyl group, etc.

(2) 2-alkyl groups: 2-ethyloctadecyl group, 2-hexyltetradecyl group, 2-octyldodecyl group, 2-decyltetradecyl group, 2-dodecylhexadecyl group, 2-tetradecyloctadecyl group, 2-hexadecyloctadecyl group, 2-tetradecyleicocyl group, 2-hexadecyleicocyl group, etc.

(3) 3-alkyl groups - 34-alkyl groups: 3-alkyl groups, 4-alkyl

groups, 5-alkyl groups, 32-alkyl groups, 33-alkyl groups, 34-alkyl groups, etc.

[0008]

Further,  $R^2$  may include mixtures of one or more branched alkyl groups, for example, mixtures of two or more groups described in the above-described items (1)-(3), propylene oligomers (from heptamer to undecamer), ethylene/propylene oligomers (molar ratio: 16/1-1/11), isobutylene oligomers (from pentamer to octamer), residues of oxoalcohols except hydroxyl groups corresponding to  $\alpha$ -olefin (containing 5-16 carbon atoms) oligomers (from dimer to heptamer), etc.

[0009]

Among  $R^2$ , preferred in regard of viscosity index and low-temperature viscosity are branched alkyl groups containing 20-32, particularly 20-28, more particularly 22-26, especially particularly 24 carbon atoms. Among these branched alkyl groups containing 24 carbon atoms, preferred are 2-alkyl groups, more preferred is 2-decyltetradecyl group.

[0010]

X is an alkylene group containing 2-4 carbon atoms and may concretely include ethylene group, 1, 2- or 1, 3-propylene group, 1, 2-, 1, 3- or 1, 4-butylene group. Among these, preferred are ethylene group and 1, 2-propylene group.

[0011]

The above-described n is 0 or an integer of 1-20, preferably 0 or an integer of 1-10, more preferably 0 or an integer of 1-5.

[0012]

5 Illustrative of the monomer (a) are 2-decyltetradecyl (meth)acrylate, 2-octyldecyloxyethyl (meth)acrylate, 2-decyltetradecyloxyethyl (meth)acrylate, 2-decylhexadecyloxypropyl (meth)acrylate, 2-dodecylhexadecyloxybutyl (meth)acrylate, etc. Among these, 10 most preferred is 2-decyltetradecyl (meth)acrylate, and a copolymer comprising this as a monomer is excellent in its viscosity-index-improvement effectiveness.

[0013]

The monomer (h) is a vinyl monomer containing one or 15 more (preferably one or two) hydroxyl groups in its molecule. Illustrative of the monomer (a) may include the following.

(h1) hydroxyalkyl (containing 2-6 carbon atoms) (meth)acrylates such as 2-hydroxyethyl (meth)acrylate, 2- or 3-hydroxypropyl (meth)acrylate, 2-hydroxybutyl 20 (meth)acrylate, 1-methyl-2-hydroxyethyl (meth)acrylate, etc.

(h2) mono- or di-hydroxyalkyl (containing 1-4 carbon atoms) -substituted (meth)acrylamides such as N,N-dihydroxymethyl (meth)acrylamide, N,N-dihydroxypropyl (meth)acrylamide, 25 N,N-di-2-hydroxybutyl (meth)acrylamide, etc.

(h3) vinyl alcohol (units thereof being obtainable by hydrolysis of units of vinyl acetate)

(h4) alkenols (containing 3-12 carbon atoms) such as (meth)allyl alcohol, crotyl alcohol, isocrotyl alcohol, 1-octenol, 1-undecenol, etc.

(h5) alkenediols (containing 4-12 carbon atoms) such as 1-butene-3-diol, 2-butene-1-ol, 2-butene-1, 4-diol, etc.

(h6) hydroxyalkyl (containing 1-6 carbon atoms) alkenyl (containing 3-10 carbon atoms) ethers such as 2-hydroxyethyl propenyl ether, etc.

(h7) aromatic monomers such as o-, m- or p- hydroxystyrene, etc.

(h8) alkenyl (containing 3-10 carbon atoms) ethers or (meth)acrylates of polyhydric (trihydric - octahydric) alcohols:

polyhydric alcohols, for instance, including alkane polyols, intramolecular or intermolecular dehydrates thereof, saccharides (for instance, glycerol, pentaerythritol, sorbitol, sorbitan, diglycerol) and sucrose;

alkenyl ethers, for example, including sucrose (meta)allyl ether

Among these, preferred in regard of viscosity index improvement is 2-hydroxyethyl methacrylate.

[0014]

The polymer (A) may further contain the following monomers (b)-(n), which can be copolymerized, other than monomers (a) and (b), as occasion demands.

[0015]

5 (b) alkyl (meth)acrylates, comprising an alkyl group containing 1-4 carbon atoms, such as, methyl (meth)acrylate, ethyl (meth)acrylate, and n-, iso- or sec-butyl (meth)acrylate

(c) alkyl (meth)acrylates (c1), comprising an  
10 (straight-chain or branched) alkyl group containing 8-18 carbon atoms, and alkyl (meth)acrylates (c2), comprising a straight-chain alkyl group containing 20-24 carbon atoms:

alkyl (meth)acrylates (c1), for instance, including  
n-octyl (meth)acrylate, 2-ethylhexyl (meth)acrylate,  
15 n-decyl (meth)acrylate, n-isodecyl (meth)acrylate,  
n-undesyl (meth)acrylate, n-dodecyl (meth)acrylate,  
2-methylundesyl (meth)acrylate, n-tridecyl (meth)acrylate,  
2-methyldodecyl (meth)acrylate, n-tetradecyl  
(meth)acrylate, 2-methyltridecyl (meth)acrylate,  
20 n-pentadecyl (meth)acrylate, 2-methyltetradecyl  
(meth)acrylate, n-hexadecyl (meth)acrylate and n-octadecyl  
(meth)acrylate;

alkyl (meth)acrylates (c2), for instance, including  
n-eicocyl (meth)acrylate, n-docosyl (meth)acrylate, etc.

25 [0016]

(d) monomers containing a nitrogen atom

(d1) monomers containing a nitro group, such as 4-nitrostyrene, etc.

(d2) vinyl monomers containing at least one of primary, secondary and tertiary amino groups, for example, including the following:

vinyl monomers containing a primary amino group, for example, including alkenyl amines containing 3-6 carbon atoms such as (meth)allyl amine, crotyl amine, etc., and aminoalkyl (having 2-6 carbon atoms) (meth)acrylates such as aminoethyl (meth)acrylate, etc.

vinyl monomers containing a secondary amino group, for example, including alkyl (containing 1-6 carbon atoms) aminoalkyl (containing 2-6 carbon atoms) (meth)acrylates such as t-butylaminoethyl methacrylate, methylaminoethyl (meth)acrylate, etc., diphenylamine (meta)acrylamides such as 4-diphenylamine (meta)acrylamide, 2-diphenylamine (meta)acrylamide, etc., and dialkenyl amines containing 6-12 carbon atoms such as di(meth)allyl amine, etc.

vinyl monomers containing a tertiary amino group, for example, including dialkyl (containing 1-4 carbon atoms) aminoalkyl (containing 2-6 carbon atoms) (meth)acrylates such as dimethylaminoethyl (meth)acrylate, diethylaminoethyl (meth)acrylate, etc., dialkyl (containing 1-4 carbon atoms) aminoalkyl (containing 2-6 carbon atoms)

(meth)acrylamides such as dimethylaminoethyl (meth)acrylamide, diethylaminoethyl (meth)acrylamide, dimethylaminopropyl (meth)acrylamide, etc.

aromatic vinyl monomers containing a tertiary amino group, such as N,N-dimethylamino styrene, etc.

vinyl monomers containing a heterocyclic having a nitrogen atom, such as morpholinoethyl (meta)acrylate, 4-vinylpyridine, 2-vinylpyridine, N-vinylpyrrole, N-vinylpyrrolidone, etc.

(d3) amphoteric vinyl monomers, for example, including N-(meth)acryloyloxy (or amino) alkyl (containing 1-10 carbon atoms) N, N-dialkyl (containing 1-5 carbon atoms) ammonium-N-alkyl (containing 1-5 carbon atoms) carboxylates (or sulfates) such as N-(meth)acryloyloxyethyl N, N-dimethylammonium N-methyl carboxylate, N-(meth)acryloyl aminopropyl N, N-dimethylammonium N-methyl carboxylate, N-(meta)acryloyloxyethyl N, N-dimethylammonium propyl sulfate, etc.

(d5) monomers containing a nitrile group such as (meth)acrylonitriles, etc.

[0017]

(e) aliphatic vinyl monomers containing a hydrocarbon, for example, including alkenes containing 2-20 carbon atoms such as ethylene, propylene, butene, pentene, heptene, diisobutylene, octene, dodecene, octadecene, etc.,

alkadienes containg 4-12 carbon atoms such as butadiene, isoprene, 1,4-pentadiene, 1,6-heptadiene, 1,7-octadiene, etc.

[0018]

- 5 (f) alicyclic vinyl monomers containing a hydrocarbon, for example, including cyclohexene, (di)cyclopentadiene, pinene, limonene, indene, vinylcyclohexene, ethylidenebicycloheptene, etc.

[0019]

- 10 (g) aromatic vinyl monomers containing a hydrocarbon, for example, including styrene,  $\alpha$ -methylstyrene, vinyltoluene, 2,4-dimethylstyrene, 4-ethylstyrene, 4-isopropylstyrene, 4-butylstyrene, 4-phenylstyrene, 4-cyclohexylstyrene, 4-benzylstyrene, 4-crotylbenzene, 2-vinylnaphthalene, etc.

15 [0020]

- (i) vinyl esters, vinyl ethers and vinyl ketones, for example, including vinyl esters of saturated fatty acids conbtaining 2-12 carbon atoms, such as vinyl acetate, vinyl propionate, vinyl butyrate, vinyl octanoate, etc., vinyl ethers of alkyls, 20 aryls or alkoxyalkyls containing 1-12 carbon atoms, such as methyl vinyl ether, ethyl vinyl ether, propyl vinyl ether, butyl vinyl ether, 2-ethylhexyl vinyl ether, phenyl vinyl ether, vinyl 2-methoxy ethyl ether, etc., and vinyl ketones of alkyls or aryls containing 1-8 carbon atoms, such as methyl 25 vinyl ketone, ethyl vinyl ketone, phenyl vinyl ketone, etc.



[0021]

(j) esters of unsaturated polycarboxylic acids, for example, including the following:

alkyl, cycloalkyl or aralkyl esters of unsaturated  
5 polycarboxylic acids, such as alkyl (containing 1-8 carbon  
atoms) diesters of dicarboxylic acids (for example, maleic  
acid, boletic acid, itaconic acid, etc.), for example,  
including dimethyl maleate, dimethyl fumarate, diethyl  
maleate, dioctyl maleate, etc.

10 [0022]

(k) vinyl monomers containing a polyoxyalkylene chain, for  
example, including the following:

polyoxyalkylene glycol (the number of carbon atoms of  
the alkylene group: 2-4, polymerization degree: 2-50), or  
15 polyoxyalkylene polyol, such as polyoxyalkylene ethers (the  
number of carbon atoms of the alkyl group: 2-4, polymerization  
degree: 2-100) of the above-described trihydric - octahydric  
alcohols

mono(meta)acrylate of alkyl (containing 1-4 carbon  
20 atoms) ethers of polyoxyalkylene glycol or polyoxyalkylene  
polyol, such as polyethylene glycol (molecular weight  
100-300) mono(meta)acrylate, polypropylene glycol  
(molecular weight: 130-500) mono(meta)acrylate, methoxy  
polyethylene glycol (molecular weight: 110-310)  
25 (meta)acrylate, lauryl alcohol ethylene oxide adduct (2-30

moles) (meta)acrylate, mono(meta)acrylic acid  
polyoxyethylene (molecular weight: 150-230) sorbitan, etc.

[0023]

(m) vinyl monomers containing a carboxyl group, for example,  
5 including the following:

unsaturated monocarboxylic acids such as (meth)acrylic  
acid,  $\alpha$ -methyl(meth)acrylic, crotonic acid, cinnamic acid,  
etc.

monoalkyl (containing 1-8 carbon atoms) ester of  
10 unsaturated dicarboxylic acids such as monoalkyl maleates,  
monoalkyl fumarates, monoalkyl itaconates, etc.

vinyl monomers containing two or more carboxyl groups  
such as maleic acid, fumaric acid, itaconic acid, citraconic  
acid, mesaconic acid, etc.

15 [0024]

Among these monomers (b), (c), (d), (e), (f), (g), (i),  
(j), (k) and (m), preferred are the monomers (b), (c) and (d),  
and using them in combination. These using in combination  
include using two or more kinds of monomers in combination,  
20 selected from the group consisting of the monomers (b), (c)  
and (d), using two or more kinds of monomers in combination,  
selected from the monomers (b), using two or more kinds of  
monomers in combination, selected from the monomers (c),  
using two or more kinds of monomers in combination, selected  
25 from the monomers (d), and their combinations.

[0025]

Of the monomers (b), preferred are monomers which comprise an alkyl group containing 1-3 carbon atoms, more preferred are monomers which comprises a methyl group. Of  
5 the monomers (c), preferred are monomers which comprise an alkyl group containing 12-18 carbon atoms.

[0026]

Of the monomers (d), preferred are the monomers (d2), more preferred are dimethylaminoethyl (meth)acrylate and  
10 diethylaminoethyl (meth)acrylate.

[0027]

The ratio of each monomer constituting the copolymer (A) is preferably decided in regard of viscosity index and low-temperature viscosity, as described below.

15 [0028]

The lower limit of the ratio of the monomer (a) is preferably 5%, (hereinafter, % expresses wt%, except when a description adds to "%".) more preferably 10%, and its upper limit is 90%, preferably 80%, more preferably 70%.

20 [0029]

The lower limit of the ratio of the monomer (h) is preferably 5%, more preferably 10%, and its upper limit of is preferably 50%, more preferably 30%, especially preferably 25%.

25 [0030]

The lower limit of the ratio of the monomers (b) is preferably 0%, more preferably 5%, and its upper limit is preferably 90%, more preferably 50%, especially preferably 40%.

5 [0031]

The lower limit of the ratio of the monomers (c) is preferably 0%, more preferably 10%, and its upper limit is preferably 75%, more preferably 60%, especially preferably 50%.

10 [0032]

In case of imparting sludge dispersibility to the copolymer (A), the monomers (d) may be used as constituent monomers, as occasion demands. In case of using the monomers (d), the lower limit of the ratio of the monomers (d), relative to the amount of all the monomers, is preferably 0.1%, more preferably 1%, especially preferably 2%, and its upper limit is preferably 10%, more preferably 7%, especially preferably 5%.

[0033]

20 The weight-average molecular weight (hereinafter referred to as Mw) of the copolymer (A) is changed in accordance with the use of the viscosity index improver. In case that the viscosity index improver is used for engine oil, the lower limit of the Mw of the copolymer (A) is preferably  
25 100,000, more preferably 150,000, especially preferably

200,000, and its upper limit is preferably 1,000,000, more preferably 600,000. In case that the viscosity index improver is used for automatic transmission fluid (hereinafter referred to as ATF), belt-continuously variable transmission fluid (hereinafter referred to as belt-CVTF) or differential gear oil, the lower limit of the Mw is preferably 5,000, more preferably 8,000, especially preferably 10,000, and its upper limit is preferably 100,000, more preferably 50,000, especially preferably 35,000, further especially preferably 20,000. In case that the viscosity index improver is used for gear oil, the lower limit of the Mw is preferably 5,000, more preferably 8,000, especially preferably 10,000, and its upper limit is preferably 100,000, more preferably 80,000, especially preferably 50,000, further especially preferably 20,000. In case that the viscosity index improver is used for hydraulic operating fluid, the lower limit of the Mw is preferably 5,000, more preferably 8,000, especially preferably 10,000, and its upper limit is preferably 400,000, more preferably 150,000, especially preferably 100,000. If the Mw is within the above-described range, shear stability suitable for each use can be imparted. The Mw is measured by gel permeation chromatography using calibration curve of polystyrene.

The Mw of the copolymer (A) can be adjusted by temperature during polymerization, the concentration of the

monomers (the concentration of solvents), the amount of catalysts or the amount of chain transfer agents.

[0034]

The lower limit of the solubility parameter  
5 (hereinafter referred to as SP value) of the copolymer (A)  
is preferably 8.6, more preferably 9.2, especially preferably  
9.4, and its upper limit is preferably 11, more preferably  
10.5, especially preferably 9.8. If the SP value is within  
the above-described range, the solubility of the copolymer  
10 (A) to base oil is further increased. The SP value is  
determined by the Fedors method [Polym.Eng.Sci.14 (2) 152,  
(1974)].

[0035]

The SP value of the copolymer (A) can be adjusted by  
15 deciding the kinds and the mole ratio of the monomers in order  
that each calculated SP value of constitution units becomes  
the target.

For example, in case that the monomers are alkyl  
(meth)acrylates, the SP value of the copolymer (A) can be  
20 adjusted by the length of an alkyl group of the alkyl  
(meth)acrylates.

[0036]

The copolymer (A) preferably has an HLB of 0.5-7,  
particularly 1-6.5, more particularly 1.5-6. If the HLB is  
25 within the range of 0.5-7, anti-emulsibility of the copolymer

(A) is further increased.

The HLB of this invention is one defined based on a concept of organic and inorganic natures ("New Introduction to Surface Active Agents" SANYO CHEMICAL INDUSTRIES, LTD.,  
5 pages 128).

[0037]

The copolymer (A) can be prepared by known polymerization methods. For instance, it is obtained by carrying out the radical polymerization of the  
10 above-described monomers within a solvent in the presence of a polymerization catalyst.

[0038]

The solvent may include, for instance, aromatic solvents such as toluene, xylene, or alkylbenzene containing  
15 9-10 carbon atoms, etc., alcohol-type solvents such as 1-propanol, 1-butanol, 2-butanol, etc., ketone-type solvents such as methyl ethyl ketone, etc., mineral oil, etc..

The, polymerization catalyst may include, for example, azo catalysts such as 2,2'-azobisisobutyronitrile,  
20 2,2'-azobis(2-methylbutyronitrile),  
2,2'-azobis-(2,4-dimethylvaleronitrile), dimethyl  
2,2'-azobisisobutyrate, etc., and peroxide catalysts such as  
t-butyl peroxy-pivalate, t-hexyl peroxy-pivalate, t-butyl  
peroxy-neoheptanoate, t-butyl peroxy-neodecanoate, t-butyl  
25 peroxy-2-ethylhexanoate, t-butyl peroxyisobutyrate, t-amyl

peroxy-2-ethylhexanoate, 1,1,3,3-tetramethylbutyl  
peroxy-2-ethylhexanoate, dibutyl peroxytrimethyladipate,  
benzoyl peroxide, cumyl peroxide, lauroyl peroxide, etc.

Furthermore, a chain transfer agent (for example, alkyl  
5 mercaptans containing 2-20 carbon atoms) can also be used as  
occasion demands. A reaction temperature is 50-140 °C ,  
preferably 60-120°C. The copolymer (A) can also be obtained  
according to bulk polymerization, emulsion polymerization,  
or suspension polymerization other than the above-described  
10 solution polymerization. Furthermore, a polymerization  
format of the copolymer (A) may include random addition  
polymerization, alternating copolymerization, Graft  
copolymerization or block copolymerization.

[0039]

15 In the invention, the monomers may be polymerized in  
the presence of a solvent to obtain the viscosity index  
improver, and the viscosity index improver may be diluted by  
a solvent to obtain an additive composition for lube oil, or  
the viscosity index improver may be diluted by a diluent after  
20 polymerization to obtain the additive composition for lube  
oil. As described above, if the additive composition for lube  
oil is previously diluted by a diluent, it is preferable that  
the additive composition for lube oil is easily dissolved into  
a lube oil when adding the former to the latter.

25 The diluent may include the following:



aliphatic solvents including aliphatic hydrocarbon containing 6-18 carbon atoms, such as hexane, heptane, cyclohexane, octane, decalin, kerosene, etc.;

aromatic solvents including aromatic solvents  
5 containing 7-15 carbon atoms, such as toluene xylene, ethylbenzene, aromatic mixed solvents containing 9 carbon atoms, such as mixtures of trimethylbenzene, ethyltoluene, etc., aromatic mixed solvents containing 10-11 carbon atoms, etc.,;

10 mineral oil such as solvent-refining oil, paraffin oil, high viscosity index oil containing isoparaffin and/or according to hydrocracking, naphthene oil;

synthetic lube oil including hydrocarbon-type synthetic lube oil such as poly- $\alpha$ -olefin-type synthetic lube  
15 oil and ester-type synthetic lube oil, etc., among these, preferred are mineral oil and synthetic lube oil, more preferred is mineral oil.

In case that the additive composition for lube oil of the present invention contain a diluent, the lower limit of  
20 the ratio of the diluent relative to the weight of the viscosity index improver of the present invention is preferably 5%, more preferably 10%, especially preferably 15%, and its upper limit is preferably 900%, more preferably 800%, especially preferably 600%.

25 If the ratio of the diluent is high, it is preferred

that the additive composition for lube oil can be dissolved into base oil easily, but if the ratio is very high, it is not economical performance.

[0040]

5           The additive composition for lube oil of the present invention may further contain, as other additives, alkyl (meth)acrylate copolymer (B) other than the copolymer (A). the copolymer (B) do not contain at least one of monomers (a) and (h) as a constitution monomer.

10       [0041]

          If the copolymer (B) is alkyl (meth)acrylate copolymers other than the copolymer (A), the copolymer (B) is not particularly limited. the copolymer (B) may include, for example, alkyl (meth)acrylates which contain an alkyl group  
15       having 1-30 carbon atoms and copolymers consisting of two or more kinds of these monomers.

          Among monomers constituting the copolymer (B), preferred are the above-described monomers (b), alkyl (meth)acrylates (q) which contain an alkyl group having 5-7  
20       carbon atoms and the above-described monomers (c). Illustrative of the alkyl (meth)acrylates (q) are n-pentyl (meth)acrylates, iso-pentyl (meth)acrylates, sec-pentyl (meth)acrylates, neopentyl (meth)acrylates, n-hexyl (meth)acrylates, n-heptyl (meth)acrylates, etc.

25       [0042]

Of the copolymer (B), preferred are copolymer (B1) comprising, as constitution monomers, one or more kinds of monomers selected from the monomers (b) and one or more kinds of monomers selected from the monomers (q) and (c), and  
5 copolymer (B2) comprising, as constitution monomers, two or more kinds of monomers selected from the monomers (q) and (c).

The ratio of the weight of the monomers constituting the copolymer (B1), that is, (the monomers (b))/(the monomers (q) + the monomers (c)) is preferably 0-40/60-100, more  
10 preferably 5-35/65-95.

The monomers of the copolymer (B2) are preferably decided, as described below. At least one of the monomers is alkyl (meth)acrylates which contain an alkyl group having 12-18 carbon atoms, and the average number of carbon atoms  
15 of the alkyl groups of all the monomers is 12-16, and the ratio of alkyl (meth)acrylates containing branched alkyl groups, relative to all the monomers, is 0-30 mol%.

Illustrative of the copolymer (B) are as follows:

the copolymer (B1) such as methyl methacrylates /  
20 n-dodecyl methacrylates / n-tetradecyl methacrylates /  
n-hexadecyl methacrylates (0-20%/20-45%/20-45%/0-20%)  
copolymers, etc.;

the copolymer (B2) such as n-dodecyl methacrylates /  
n-hexadecyl methacrylates (10-50%/50-90%) copolymers (the  
25 average number of carbon atoms: 12.3-13.8, the ratio of

branched alkyl groups: 0 mol%], n-dodecyl methacrylates /  
n-tetradecyl methacrylates (90-70%/10-30%) copolymers (the  
average number of carbon atoms: 12.2-12.6, the ratio of  
branched alkyl groups: 0 mol%], n-dodecyl acrylates /  
5 n-tetradecyl methacrylates (10-40%/90-60%) copolymers (the  
average number of carbon atoms: 12, the ratio of branched  
alkyl groups: 0 mol%), etc.

As the copolymer (B), only one kind of copolymer  
selected from the above-described copolymers may be used, and  
10 two or more kinds of copolymers selected from the  
above-described copolymers may be used in combination.

The Mw of the copolymer (B) is preferably  
5,000-1,000,000, more preferably 10,000-250,000.

In case of using the copolymer (B), the ratio of the  
15 copolymer (B), relative to the weight of the copolymer (A),  
is preferably 0-30%, more preferably 0.001-20%, especially  
preferably 0.001-10%. In case of the copolymer (B) consisting  
of only the copolymer (B1), the ratio of the copolymer (B),  
relative to the weight of the copolymer (A), is preferably  
20 0-30%, more preferably 0.001-25%, especially preferably  
0.001-20%, above all especially preferably 0.001-10%. In  
case of the copolymer (B) consisting of only the copolymer  
(B2), the ratio of the copolymer (B), relative to the weight  
of the copolymer (A), is preferably 0-15%, more preferably  
25 0.001-10%, especially preferably 0.001-8%. If the ratio is

within above-described range, a desirable low-temperature viscosity can be achieved.

[0043]

Each of other additives of the present invention may  
5 be not included in the additive composition for lube oil and  
be solely added to the base oil, or may be solely dissolved  
into a diluent, to prepare solution added to the base oil.  
For example, a solution comprising the copolymer (B) and  
diluent, and the additive composition for lube oil of the  
10 present invention, comprising the copolymer (A) and a diluent,  
may be added to the base oil respectively.

[0044]

Base oil, into which the viscosity index improver of  
the present invention is added, is not particularly limited.

15 The base oil preferably have a kinematic viscosity of  
1-18 mm<sup>2</sup>/s, preferably 2-15 mm<sup>2</sup>/s, at 100°C, and a viscosity  
index of preferably at least 60, more preferably at least 100,  
especially preferably 105-180. If a lube oil composition  
comprises such base oil and the viscosity index improver of  
20 the present invention, further high viscosity index and  
preferable fuel-efficiency can be achieved.

[0045]

The base oil, into which the viscosity index improver  
of the present invention is added, preferably have a pour  
25 point (defined in JIS K2269-1993) of -5 °C or less,

particularly  $-10^{\circ}\text{C}$ -- $-70^{\circ}\text{C}$ . If the pour point is within the above-described range, the amount of wax deposition is decreased and low-temperature viscosity is preferable. The base oil may include, for example, solvent-refining oil, high  
5 viscosity index oil containing isoparaffin and/or according to hydrocracking, hydrocarbon-type synthetic lube oil (poly- $\alpha$ -olefin-type synthetic lube oil), ester-type synthetic lube oil, naphthene oil, etc. They may be used solely, or two or more kinds of base oils selected from them may be used  
10 in combination.

[0046]

The lube oil composition is prepared by adding the viscosity index improver comprising the copolymer (A) of the present invention in order that the ratio of copolymers (A)  
15 relative to the total weight of the lube oil composition is preferably 0.5-60%, more preferably 5-50%, especially preferably 12-45%.

[0047]

In case that the lube oil composition is used as engine  
20 oil, preferably, base oil having a kinematic viscosity of 3-10  $\text{mm}^2/\text{s}$  at  $100^{\circ}\text{C}$  is used and is added in order that the ratio of the copolymer (A) is 0.5-15%, relative to all the weight of the lube oil composition.

In case that the lube oil composition is used as  
25 automatic transmission fluid (ATF, belt-CVTF), preferably,

base oil having a kinematic viscosity of 2-6 mm<sup>2</sup>/s at 100°C is used and is added in order that the ratio of the copolymer (A) is 2-35%, relative to all the weight of the lube oil composition.

5           In case that the lube oil composition is used as gear oil or differential gear oil, preferably, base oil having a kinematic viscosity of 3-18 mm<sup>2</sup>/s at 100°C is used and is added in order that the ratio of the copolymer (A) is 2-40%, relative to all the weight of the lube oil composition.

10           In case that the lube oil composition is used as traction oil, preferably, base oil having a kinematic viscosity of 1-5 mm<sup>2</sup>/s at 100°C is used and is added in order that the ratio of the copolymer (A) is 0.5-15%, relative to all the weight of the lube oil composition.

15           In case that the lube oil composition is used as hydraulic operating fluid, preferably, base oil having a kinematic viscosity of 1-10 mm<sup>2</sup>/s at 100°C is used and the base oil is added in order that the ratio of the copolymer (A) is 0.5-25%, relative to all the weight of the lube oil composition.

20

[0048]

          In case of using the copolymer (B), the copolymer (B) is preferably added in order that the ratio of total amount of the copolymers (A) and (B) is preferably 1-50%, more preferably 5-45%, relative to all the weight of the lube oil

25

composition.

[0049]

The lube oil composition of the present invention may contain other optional additives, for example:

5       detergents (for instance, calcium or magnesium salts such as sulfonates, sulfonates, phenates, naphthenates, etc., carbonates) in an amount of 0-20%, preferably 0.1-10%, relative to the weight of the copolymer (A);

10       dispersants such as instance, succinimides (for example, bis-, mono- or borate-type, Mannich condensation products, etc.) in an amount of 0-20%, preferably 0.2-10%;

15       anti-oxidants such as zinc dithiophosphates, and bamines (for example, diphenylamine, hindered phenols, zinc thiophosphate, trialkylphenol, etc.) in an amount of 0-5%, preferably 0.1-3%;

20       Oiliness improvers (for instance, long chain fatty acids such as oleic acid, esters of long chain fatty acids such as oleates, long chain amines such as oleylamine, long chain amides such as oleylamide) in an amount of 0-5%, preferably 0.1-1%;

      friction modifiers such as molybdenum dithiophosphate, molybdenum carbamate, zinc dialkyl dithiophosphate, etc. in an amount of 0-5%, preferably 0.1-3%;

25       extreme-pressure agents such as sulfur compounds, sulfur phosphide compounds, phosphide compounds, chlorine



compounds, etc. in an amount of 0-20%, preferably 0.1-10%;

antifoam agents such as silicone oils, metallic soaps, fatty acid esters, phosphates, etc. in an amount of 2-1000ppm, preferably 10-700ppm;

5 demulsifying agents such as quaternary ammonium salts, sulfated oils, phosphates in an amount of 0-3%, preferably 0-1%; and

corrosion inhibitors (for instance, nitrogen compounds such as benzotriazole, sulfur and nitrogen-containing  
10 compounds such as 1,3,4-thiodiazoryl-2,5-bisdialkyl dithiocarbamate, etc.) in an amount of 0-3%, preferably 0-2%.  
[0050]

The lube oil composition of the present invention has an excellent shear stability, and in case of testing it  
15 according to CEC L45-45-A-99 for test period of 20 hours, the ratio of a decrease in viscosity is preferably 20% or less, more preferably 15% or less, especially preferably 10% or less, most preferably 5% or less.

[0051]

20 The lube oil composition of the present invention is used as engine oil, which is used in an engine such as an engine for means of transportation and engine for machine tools, gear oil, differential oil, automatic-transmission oil (ATF, CVT oil), traction oil, shock-absorber oil, power-steering oil,  
25 hydraulic operating fluid, etc. Among these, preferable uses

are gear oil, differential oil, automatic-transmission oil, hydraulic operating fluid. Especially preferable uses are differential oil and gear oil.

[0052]

5 Hereinafter, the invention will be described more particularly according to examples, however the invention is not at all restricted to these examples. In the examples and comparative examples, parts and % are by weight.

[Measurement method of Mw by GPC]

10 Apparatus: HLC-802A, produced by Tosoh Corp.

Column: TSKgel-GMH6, two columns

Temperature: 40°C

Sample solution: 0.5 wt% of tetrahydrofuran solution

Injection amount: 200  $\mu$ l

15 Detector: refractive index detector

Standard: polystyrene

[0053]

[Test method of low-temperature viscosity]

The viscosity was measured at -40 °C according to  
20 JPI-5S-26-85.

[0054]

[Test method of viscosity index]

The viscosity index was measured in accordance with  
JIS-K-2283.

25 [0055]

[Test method of shear stability]

The shear stability was measured according to CEC-L45-45-A-99 for test period of 12 hours.

[0056]

5 Example 1 and comparative example 1

25 parts of toluene was put into a reaction vessel equipped with a stirrer, a heating and cooling device, a thermometer, a dropping funnel and a nitrogen inlet tube. 100 parts in total of monomers described in Table 1, 1.5 parts of dodecylmercaptan as a chain transfer agent, and 0.5 parts of 2,2'-azobis-(2,4-dimethylvaleronitrile) (hereinafter referred to as ADVN) as a radical polymerization initiator were put into other glass beaker, and were mixed by stirring at 20°C to prepare monomers solutions, and each monomers solution was put into the dropping funnel respectively. After a gas phase part of the reactor was substituted with nitrogen gas and sealed, while maintaining the temperature within 70-85°C, each monomers solution was added thereto dropwise over 2 hours respectively, and was aged at 85°C for two hours after adding to obtain a polymer respectively. Toluene was distilled out from each polymer at 130°C for 3 hours under reduced pressure to obtain copolymers (A-1) and (X-1) as viscosity index improvers. The properties of the copolymers are shown in Table 1.

25 [0057]

[Table 1]

	Copolymer	Monomer (parts)				
		a-1	h-1	b-1	c-1	c-2
Example 1	A-1	50	19	-	31	-
Comparative Example 1	X-1	-	-	18	80	2

Codes of monomers:

a-1: 2-decyltetradecyl methacrylate

h-1: 2-hydroxyethyl methacrylate

5 b-1: methyl methacrylate

c-1: n-tetradecyl methacrylate

c-2: n-octadecyl (meth)acrylate

[0058]

[Table 2]

	Copolymer	Mw	SP value	HLB value
Example 1	A-1	$2 \times 10^4$	9.6	3.3
Comparative Example 1	X-1	$2 \times 10^4$	9.1	2.5

10 [0059]

Example 2 and comparative example 2

65 parts of the copolymer (A-1) or (X-1) was respectively mixed with 35 parts of mineral oil (kinematic viscosity:  $2.3 \text{ mm}^2/\text{s}$  at  $100^\circ\text{C}$ ), and was dissolved into the  
 15 mineral oil to prepare additive composition for lube oil respectively.

[0060]

### Example 3 and comparative example 3

The additive composition for lube oil containing the copolymer (A-1) or (X-1), and base oil (kinematic viscosity: 4.6 mm<sup>2</sup>/s at 100°C, viscosity index: 118, pour point: -17.5°C) were added into a stainless container equipped with a stirring mixing device to prepare a lube oil composition of the present invention or comparative example, in order that the lube oil composition has a kinematic viscosity of 14.3±0.2 mm<sup>2</sup>/s, and its total amount is 100 parts. In Table 6 shown are kinematic viscosity at 100 °C, viscosity index, low-temperature viscosity at -40 °C and shear stability of the lube oil compositions.

[0061]

[Table 3]

	Copolymer	Additive composition for lube oil (Mount of addition)	Kinematic viscosity at 100°C (mm <sup>2</sup> /s)	Viscosity index	Viscosity at -40°C (mPa · s)	shear stability (%)
Example 1	A-1	45	14.3	223	61,000	3.5
Comparative Example 1	X-1	45	14.3	175	400,000 or more	7

15 [0062]

[Effect of the Invention]

As compared with the conventional lube oil composition containing polymethacrylate-type viscosity index improver,

the lube oil composition containing the viscosity index improver of the invention can improve viscosity index, and is also excellent in shear stability and low-temperature viscosity, therefore, manual transmission gear oil  
5 corresponding to the fuel-efficiency of future automobiles and SAE-J306 can be produced without using mineral oil. Accordingly, it can be used preferably as drive-system lube oil (for example, manual transmission oil, differential gear oil, automatic-transmission oil, belt-CVT oil, etc.),  
10 hydraulic operating oil (for example, hydraulic oil of machines, power-steering oil, shock-absorber oil, etc.), engine oil (engine oil for gasoline engines, diesel engines, etc.), and traction oil.

[Name of Document] Abstract

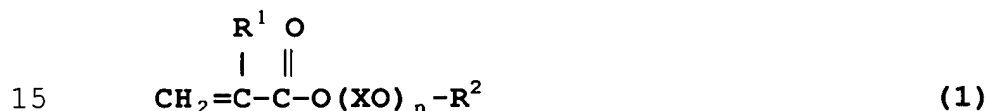
[Abstract]

[Subject] To provide a viscosity index improver excellent in  
viscosity-index-improvement ability and a lube oil  
5 composition containing it.

[Means of solving] A viscosity index improver, which  
comprises a copolymer (A) comprising, as essential  
constituent monomers, a monomer (a) represented by the  
following general formula (1) and a monomer (h) which contains  
10 a hydroxyl group:

General formula

[Chemical formula 3]



wherein  $\text{R}^1$  is a hydrogen atom or a methyl group, X is an  
alkylene group containing 2-4 carbon atoms, n is 0 or an  
integer of 1-20, and  $\text{R}^2$  is a branched alkyl group containing  
20-36 carbon atoms.

20 [Selected Fig.] Nil